Bevalac Decommissioning

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Following a long and distinguished scientific career, the Bevalac complex was closed in 1993. Because of community sensitivity and legal requirements for the disposal of radioactive materials, it is critical that the characterization of man-added radionuclides be done and documented as thoroughly as possible. The social and legal consequences of less-than-complete characterization are extensive, as evidenced from the difficulties experienced by LBNL with citizen opposition to the tritium facility and its decommissioning.

While extensive and complete monitoring of all samples leaving the building is necessary, it is not as difficult as it may sound.

Central to this process is the use of one or more Ge spectrometers to identify with certainty the gamma-ray emitting radionuclides, mainly in the concrete and steel components (95% or more of the total mass to be disposed of). The suite of radionuclides expected is relatively small: both steel and concrete contain trace amounts of Co-59 which activates to Co-60 (5.3 yr. half-life) via the capture of slow neutrons. Similarly, trace amounts of natural Eu in normal density concrete and the large amount of Ba in some of the high density concrete (other high-density concrete contains Fe) activate to Eu-152 (13.3-yr. half-life) and Cs-137 (30-yr. half-life) respectively. High-energy reactions on steel, copper, and brass can produce Na-22 (2.6-yr. half-life), Ti-44 (67-yr. half-life) and Ar-42 (33-yr. half-life). Typically, runs of 10 minutes can determine if any of these isotopes are in the samples at the pico-Curie/g level. Noting that natural normal density concrete typically contains 10-20 pico-Curies/g of the naturally–occurring isotopes of U, Th, and K, sensitivity to man-added activity at a level of better than 10% of natural activities (considered harmless) are achieved.

While the problem of identifying radioisotopes is simple with this instrument, the problem of quantifying the amounts is more difficult, requiring detailed knowledge of the distribution of activity within the sample as well as the energy-dependent detector efficiency.

Several things work in our favor in this regard. First is that for large articles, such as concrete blocks or magnet steel, the distribution of activity usually peaks at the surface and falls off as one penetrates the material. Second, absorption in the bulk material evens out the energy dependence of the detector efficiency, making it nearly constant over the energy range 300-3000 keV. Thus, as shown in reference 1, a simple relationship exists between the counts observed and the activity in the bulk material. For example, for Co-60, the observed counts per minute in one of the characteristic spectral lines divided by 35 gives activity in pico-Curies/g. Similarly, and of importance where odd-shaped items or items where the inside or restricted access parts need to be analyzed, other meters sensitive to micro-R/hr can be used. A reading of 17 micro-R/hr corresponds to an activity of 10 pico-Curies/g of Co-60.

This combined approach, using the resolution of the Ge spectrometer to identify (and in some cases to quantify) activity; and in some cases to use the Ge spectrometer only for identification and other instruments for quantification, will provide complete but cost-effective characterization. In many cases, where obvious activity is observed, and the material will need to disposed of a radioactive, the precision of activity measurement required will only be within the wide range allowed for the particular disposal method.

In conclusion, it is critical that the radioactivity characterization of Bevalac components slated for disposal or reuse is done in a comprehensive manner to limit LBNLs exposure to negative public opinion or legal action. Critical to this is the use of a Ge spectrometer to identify (and quantify) activity in the materials.

Ref. 1 R.J. McDonald, A.R. Smith, D.L. Hurley, E.B. Norman, and M.R. Schoonover. J. Radioanalytical and Nuclear Chem. V-233 (1998) 191-194

Homeland Security and Automotive Air Filters

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Following the 9/11 destruction of the World Trace Center Buildings and the subsequent anthrax attack on the Hart Senate Office Building, Homeland Security has become both a national and a personal priority. No one knows when or where the next attack will come, and no one knows the nature of the attack. Besides explosives, chemical, biological, or nuclear materials could be involved.

Upon notification that the Chernobyl accident had spread fission products over large parts of Europe, the LBNL Environmental Health and Safety Department set up a special on-site air sampling station. Sampling times varied from 6 to 24 hours, after which filters were immediately brought to the Low-Background Facility (LBF) for detailed gamma-ray spectrographic analysis.

Similarly, the LBF performed the same kind of analysis on a set of 14 environmental air filters on the LBNL site and an additional site at the Oroville Dam, 180 miles northeast of Berkeley. These sets of filters provided normal background information, including the naturally occurring Be-7 (from cosmic ray interactions in the upper atmosphere) and Pb-210 (from the decay of airborne Rn-222).

Chernobyl debris first appeared on LBNL air filters on 5/9/86 with the appearance of the short-lived nuclides Te-123, I-131, and Ru-103 as well as the long lived Cs-134 and Cs-137. The concentrations observed were, fortunately, small compared to levels of public health concern. This sampling program continued through June 1986, by which time the fallout from Chernobyl declined below detectability.

Recognizing the value of air filters as collectors of fallout; several engine air filters from motor pool cars were analyzed for radioactivity. The car used for the Berkeley-Oroville trip on 5/9 and 5/10 1986 showed 20-25% of the activity of diagnostic nuclides observed at the dedicated LBNL air collection system. The value of automotive air filters as collectors was also recognized in Europe in the aftermath of the Chernobyl accident.

The essence of this experience is that an ubiquitous item from our automotive culture, the intake air filter, can be used as a collector of airborne contamination in the form of chemical, biological, or radiological hazard. At any time, over 100 million such collectors are continuously deployed in a pattern that mimics population distribution and movement. Following an attack, these collectors may be examined for contamination.

The LBF program is to determine how quantitative is the method via a pilot program involving sets of filters obtained from police vehicles from Oroville and Berkeley as well as filters obtained from a local repair shop. The purpose of this is to obtain baseline parameters utilizing the naturally occurring nuclides Be-7 and Pb-210.

Following demonstration of the principle and determination of the background (and time dependent) parameters, a nation wide surveillance system can be established. Local law enforcement vehicles constitute the most readily available set of collectors, and filters could be forwarded to local analysis centers, located at universities, hospitals, reactors, or other sites with access to Ge spectrometers. These sites must be identified beforehand and they must be supplied with some calibration standard, for example potassium chloride.

The facilities and experience at the LBF makes this LBNL facility the proper lead organization to demonstrate and coordinate an effort that should become nation wide to make this 100 million collection sites available and useful in the case of an attack involving radioactivity.

Additional LBNL studies should be added to establish the suitability of this method to detect chemical and biological agents.

Low-background Counting Facilities

A.R. Smith, D.L. Hurley , R.J. McDonald, and E.B. Norman

The LBNL Low Background Facilities (LBF) consist of a Berkeley site and an Oroville site specially configured for low-background gamma-ray spectroscopy. The Berkeley site was established in 1963 and consists of a 3m by 7m x 3m room surrounded by 1.6m of specially selected low-background concrete shielding. The aggregate in this concrete is from serpentine gravel, which is low in U, Th, and K, and emits very little radon.

Detectors at this site include a 20 cm diameter by 10 cm thick NaI crystal, two 30% p-type Ge spectrometers, two 80% p-type Ge spectrometers, available for fieldwork, and a 115% n-type spectrometer suitable for observing Pb-210 decay. These detectors each have small local shields consisting of 10 cm of Pb. The overall shielding reduces background to the point where cosmic rays and activity within the detector assembly are the dominant sources of background.

The LBF Oroville site is located in the powerhouse of the Oroville Dam, under 180-m of rock cover. This site now has an 80% p-type Ge-spectrometer, and is used for our most sensitive counting, particularly for materials certification. Sensitivities of 50 parts-per-trillion (PPT) for U and daughters, 200 PPT for Th and daughters, and 100 parts-per-billion for K are realized at the Oroville site.

Over the years, the LBF has been involved in a wide variety of experiments supporting programs in basic and applied science from LBNL and a variety of other institutions. This last year, work mainly involved: 1) low-activity materials certification for CDMS and KamLAND, 2) various activities related to CUORE (Cryogenic Underground Observatory for Rare Events), 3) Bevalac Decommissioning, and 4) use of automotive air cleaners as collectors for radioactive aerosols following a potential nuclear terrorist act. 5) studies of radioactivity in and around CCDs used in astronomy applications, and 6) site characterization of natural and fallout radionuclides in surficial soils.

Certification of materials work continued for CDMS and KamLAND, mostly involving direct counting. Some of the CDMS work involved neutron activation of materials at the McClellan Nuclear Research Center in California.

Neutron activation work continued with the fabrication of NDT Ge thermistors for the CUORE experiment in Italy. Irradiations were performed at both the MIT and Missouri reactors. Protocols are being developed to produce the one-to two-thousand nearly identical thermistors needed for CUORE.

A large amount of work went into the actual construction of the improved MiBeta and Cuoricino (little CUORE) experiments in Italy, including crystal polishing, thermistor attachment, and tower construction.

Work has begun on Bevalac decommissioning. Current studies involve counting both concrete and steel components in order to determine the magnitude of the disposal project. Samples of nuts and washers around the ring were counted to map out the radioactivity in the steel. 110 large concrete blocks were counted and certified clean for free release. Current efforts involve counting magnets and steel for disposal.

A new project related to Homeland Security was started to study radioactivity trapped in automotive air filters. The sensitivity to Co-60 is better than 100 pico-Curies total activity on the filter, as determined from the natural Be-7 activity. This technique could be used to monitor the spread of radioactivity following a terrorist attack. Air filters from Berkeley and Oroville are being studied.

Large CCD arrays for astronomy show spurious events not related to observation or cosmic rays. Studies at the Oroville site confirm that these events are from radioactivity in materials near the CCDs, including activity in the concrete of the telescope. We are in the process of studying these CCDs with neutron activation.

CUORE (Cryogenic Underground Observatory for Rare Events)

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CUORE (Cryogenic Underground Observatory for Rare Events) is designed to be a large (750kg) array of 1000 TeO_2 crystals to search for rare events, particularly for the neutrinoless double beta decay of ^{130}Te. This experiment will succeed the recently completed MiBeta experiment and the Cuoricino experiment (62 crystals) under construction.

Work in three major areas proceeded for this experiment: 1) construction of the last tests of the MiBeta experiment, 2) crystal polishing for Cuoricino, and 3) continued work on NTD Ge thermistors.

One of us (McDonald) spent a total of 9 weeks at the Laboratorio Nazionale del Gran Sasso working on the final tests utilizing the MiBeta apparatus, an array of 20 3x3x6 cm TeO2 crystals. The goal of this test was to determine if background could be reduced by using ultra-clean copper in the assembly and by lapping away 30 microns of surface to reduce observed surface radioactivity. We also designed a borated polyethylene neutron shield predicted to reduce neutron-induced background by about 30%. These tests have now been completed, and results are being analyzed.

Two of us (Norman and McDonald) spent a total of 9 man-weeks at the Universita' dell' Insubria in Como polishing crystals for the Cuoricino experiment. A variety of hand and machine polishing techniques were tried. As delivered from Shanghai, the crystals were not lapped to the extent expected, and the process of rough lapping had to be repeated. Final lapping was done by hand on two different polishing cloths, resulting in uniformly opaque surfaces. Tests at the Gran Sasso indicated that optically clear surfaces were not required for adequate detector performance.

The thousands of nearly identical thermistors needed CUORE will be produced by the neutron transmutation doping (NTD) of natural germanium. This involves subjecting Ge to about 3×10^{18} neutrons to produce doping levels of about 1×10^{17} Ga atoms per cm^3, 3×10^{16} As atoms per cm^3 and 2×10^{15} Se atoms per cm^3. The word "about" as used here implies only that the absolute numbers are not well determined, whereas precision on the order of 1% will be required to produce thermistors with characteristics close enough to be used.

Our efforts have mainly been focused on studying monitor foils to determine the thermal and epithermal flux in the reactor and the predicted production of dopants. The goal is to use monitor foils as proxies for the production of dopants since the electrical properties of the thermistors cannot be measured for one year or more after the irradiations because of the 11.2 day half life of Ge-71.

One test involved irradiations at the MIT research reactor. This reactor has the advantage of a D2O moderator that essentially eliminates the epithermal portion of the neutron spectrum. Foils of Cr, Fe, and Zr confirmed the absence of epithermal neutrons. Further tests need to be performed to find a part of the reactor with constant and uniform neutron flux to produce large numbers of identical thermistors.

A second test involved increasing the doping of Ge originally irradiated two years ago. Unfortunately, Zr foils were not used at that time, and there was some uncertainty about the neutron flux and spectrum. The additional irradiation was aimed to span the predicted doping. These samples are not sufficiently decayed to test their electrical properties.

Throughout these tests, we continue to build experience that will lead to the protocols for producing the numbers of thermistors needed for CUORE.

Once Cuoricino is taking data, we expect to prepare a full proposal for CUORE in partnership with the Milano Group.

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